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Radiation Dose Estimates Due to Air Particulate Emissions from Selected Phosphate Industry Operations

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U.S. ENVIRONMENTAL PROTECTION AGENCY

Office of Radiation Programs

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Technical Note
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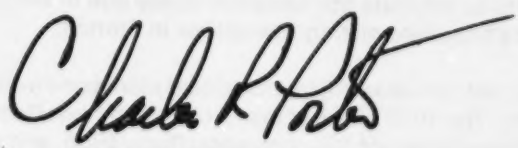
This report has been reviewed by the Environmental Protection Agency (EPA) and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the EPA, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

PREFACE

The Eastern Environmental Radiation Facility (EERF) participates in the identification of solutions to problem areas as defined by the Office of Radiation Programs. The Facility provides analytical capability for evaluation and assessment of radiation sources through environmental studies and surveillance and analysis. The EERF provides technical assistance to the State and local health departments in their radiological health programs and provides special analytical support for Environmental Protection Agency Regional Offices and other federal government agencies as requested.

This study is one of several current projects which the EERF is conducting to assess environmental radiation contributions from naturally occurring radioactivity.



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ABSTRACT

The EPA Office of Radiation Programs has conducted a series of studies to determine the radiological impact of the phosphate mining and milling industry. This report describes the efforts to estimate the radiation doses due to airborne emissions of particulates from selected phosphate milling operations in Florida.

Two "wet process" phosphoric acid plants and one ore drying facility were selected for this study. The 1976 Annual Operations/Emissions Report, submitted by each facility to the Florida Department of Environmental Regulation, and a field survey trip by EPA personnel to each facility were used to develop data for dose calculations. The field survey trip included sampling for stack emissions and ambient air samples collected in the general vicinity of each plant. Population and individual radiation dose estimates are made based on these sources of data.

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I. Introduction

The EPA Office of Radiation Programs has been conducting studies of the radiological impact of the phosphate mining and milling industry.(1-4) Phosphate ore has been shown to contain varying amounts of naturally occurring radionuclides of ^{238}U and ^{232}Th series (figure 1). The mining and milling of these ores results in the dispersal of radium, uranium, thorium, and other radionuclides throughout the environment, which could increase the radiation doses to the general population. The objective of this investigation was to estimate the population and individual radiation doses due to airborne emissions of particulates around selected phosphate milling and processing facilities in central Florida.

II. General Processes: Description and Emissions

A. Phosphate Rock Processing

The preparation of phosphate rock generally involves strip mining to obtain ore, benefication to remove impurities, drying to remove moisture, and grinding to improve reactivity. These operations are shown graphically in figure 2. In the strip mining operations the overburden is stripped from above the phosphate ore using electric draglines. The ore is removed by the same dragline and dropped into a sluice pit. In this pit, high pressure water is used to produce a slurry which is then pumped to the washer plant.

In the washing and benefication process, marketable rock is separated from sand tailings and clay slimes. This is accomplished through a series of screening and flotation steps.

From the washing process the marketable rock is transferred to the drying and storage area. Here the wet rock is dried in large rotating drums. After drying, the rock is separated according to size and grade and stored. The material from the dryers may be ground using ball mills before storage.

The predominate airborne emissions from this portion of the phosphate industry are in the form of fine rock dust from drying and grinding operations. Phosphate rock dryers are usually equipped with dry cyclones followed by wet scrubbers. Phosphate rock grinders can be a considerable source of particulates. Because of the extremely fine particle size, baghouse collectors are normally used to reduce emissions.

B. Phosphoric Acid

Phosphoric acid is produced by two principal methods, the wet process and the thermal process. The wet process is usually employed when the acid is to be used for fertilizer production, and the thermal process is normally used for high-grade chemical and food products.

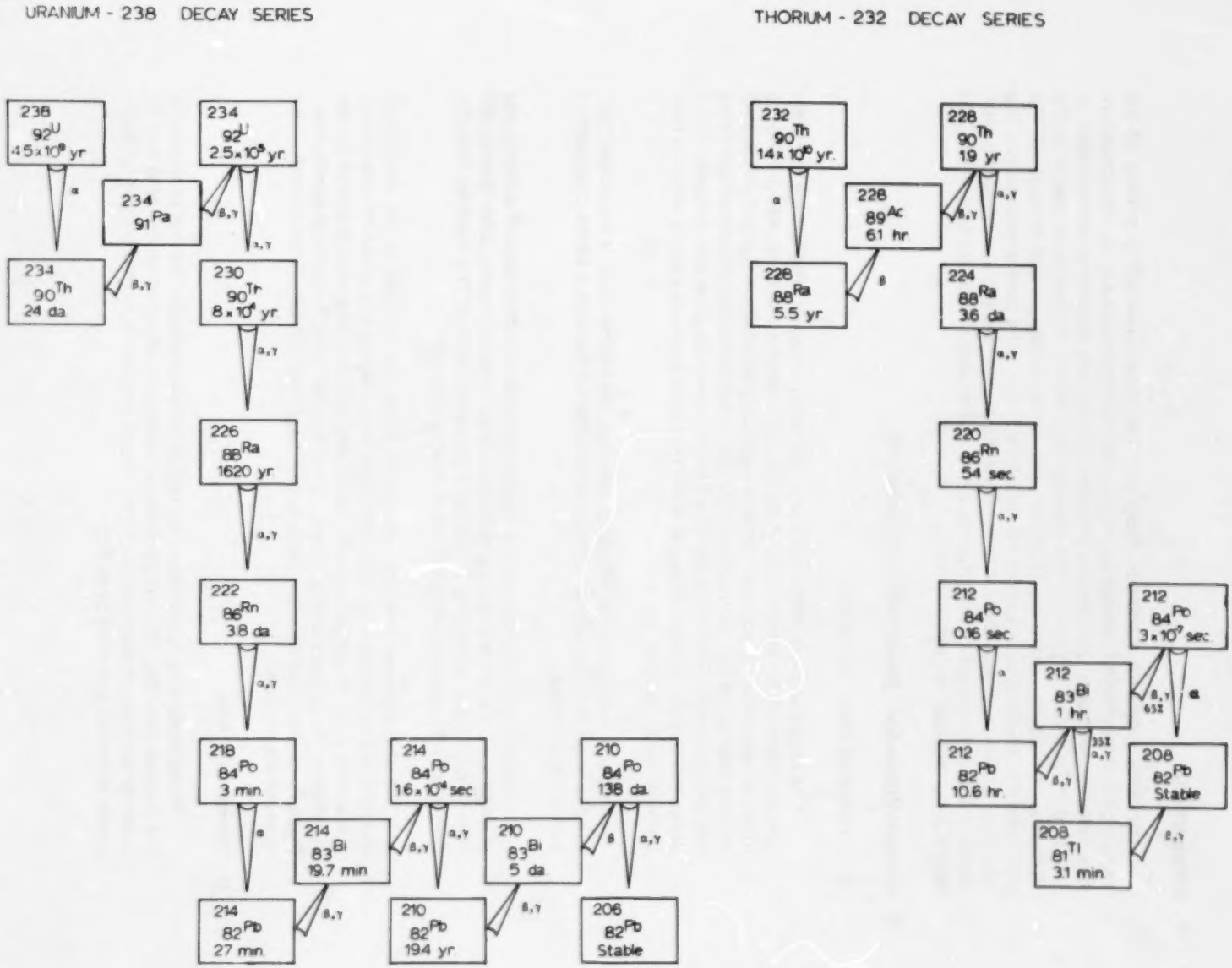

Figure 1. ^{238}U and ^{232}Th decay series.

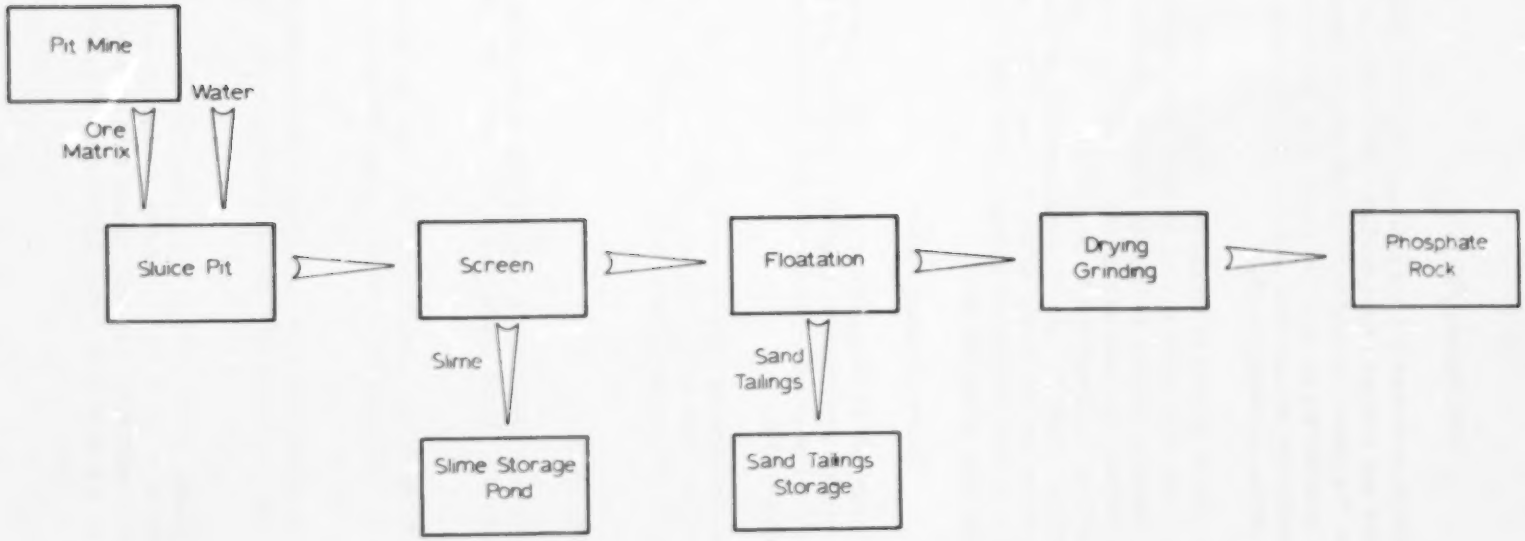

Figure 2. Phosphate rock processing, flow diagram.

Wet Process Plants

The general operations performed at a wet process phosphoric acid plant are shown in figure 3. In the wet process plants studied, phosphate rock is usually received in railroad cars. The rock is dropped from car hoppers onto conveyor belts which moves the rock to temporary storage. Car vibrators or shakers are used to help dislodge rock packed in the railroad cars causing a very dusty environment in the immediate vicinity of the unloading facility.

From storage, the rock is ground as necessary, using large ball mills. The crushed rock is then mixed with sulfuric acid in the reactor vessel to make phosphoric acid. After reaction, calcium sulfate (gypsum) is separated from the phosphoric acid in pan filters and pumped as a slurry to the waste gypsum pile. The phosphoric acid is normally concentrated to 54 percent P_2O_5 and is then transferred to the fertilizer plant for use in manufacturing various fertilizer products. Gaseous fluorides are the major airborne emission problem in wet process phosphoric acid facilities. Additional emission problems result from the transferring of phosphate rock within the plant.

Thermal Process Plants

In the "thermal process" plants phosphate rock, coke and siliceous material are electrically smelted in a furnace (figure 4). Elemental phosphorus is recovered by condensing vapors from the furnace. The elemental phosphorus can then be used to produce phosphoric acid.

Although elemental phosphorus is the principal product at these facilities, ferrophosphorus and slag are also sold for various uses. The major atmospheric contaminant from thermal process phosphoric acid manufacture is phosphoric acid mist.

C. Phosphate Fertilizer Production

Phosphatic fertilizers can generally be divided into three categories: *(1) normal superphosphate, (2) triple superphosphate, and (3) ammonium phosphates.*

Normal superphosphate is the product resulting from the acidulation of phosphate rock with sulfuric acid. Normal superphosphate contains from 16 to 22 percent P_2O_5 .

Triple superphosphate is the product of the reaction between phosphate rock and phosphoric acid. The product usually contains approximately 46 percent P_2O_5 .

The two general classes of ammonium phosphates are monoammonium phosphate and diammonium phosphate. Several processes are used to manufacture ammonium phosphates. Basically, phosphoric acid, sulfuric acid, and anhydrous ammonia are allowed to react to produce the desired grade of ammonium phosphate.

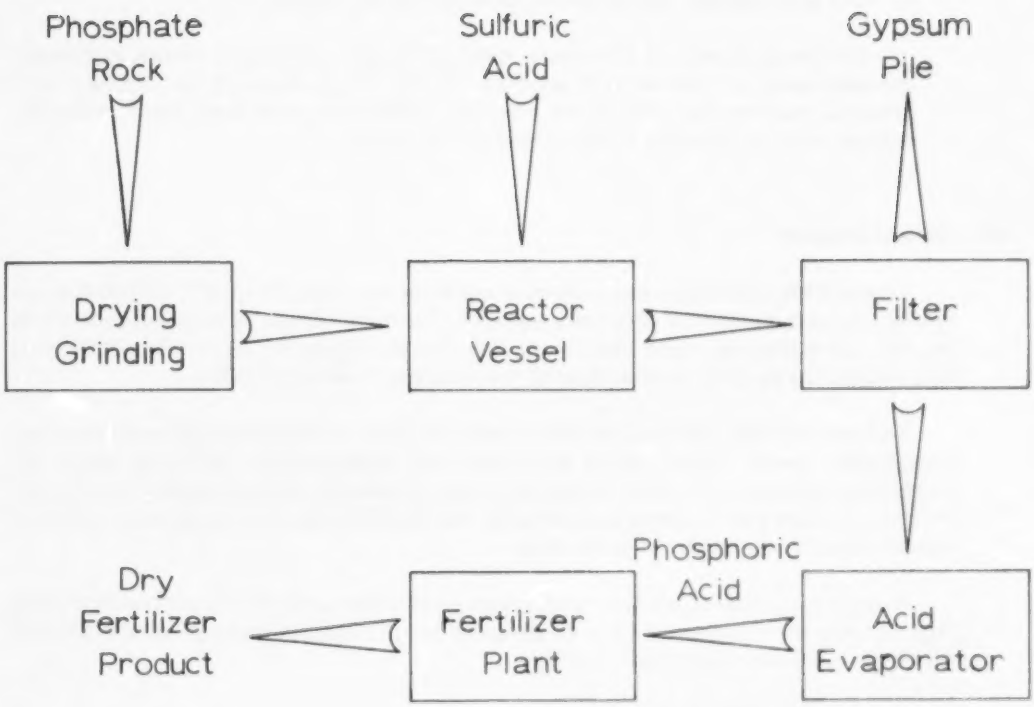

Figure 3. Wet process phosphoric acid flow diagram.

The primary airborne emissions from the production of phosphate fertilizers are gaseous fluorides and some particulates from the grinding, drying, and storage of the products.

Most facilities have granular fertilizer stored in large warehouse buildings. The product is deposited in the building via conveyer belts and from there it is outloaded as required using front-end loaders. When the product is being moved by front-end loaders, the airborne particulates are visible.

For the purposes of this study, only particulate emissions known to contain elevated levels of radioactivity were of interest. Those sources, determined from previous studies, are operations involving phosphate rock dust and the drying, storing, and shipping of finished fertilizer products.

III. Data Collection

To estimate population and individual radiation doses in the vicinity of the selected operations, two sources of data were used: *(1) the 1976 Annual Operations/Emissions Report submitted by each facility to the Florida Department of Environmental Regulation; (2) an EPA field survey of the facilities (February 1977).*

Two wet process phosphoric acid plants and one ore drying facility were selected for detailed study. These plants were selected because they generally typify the phosphate industry in central Florida. Only wet process phosphoric acid plants (i.e. no thermal process plants) were evaluated in this study since this is the most common type of plant in the central Florida area.

In addition to these facility oriented data, ambient air samples were collected using high-volume air samplers. These air samples were collected throughout the general area of phosphate mining and milling.

A. Facility Reported Data

Each of three facilities studied supplied a copy of their 1976 Annual Operations/Emissions Report. These reports detail total particulates given in average pounds per hour and in total tons per year for each source. From these reports total particulate emissions from sources known to contain radioactivity were determined.

Annual airborne particulate emissions for each facility are summarized in tables 1, 2, and 3. Radioactivity emissions were calculated by assuming the concentration of radioactivity in the effluent from a given process to be the same as the concentration in the raw products. Following this assumption, total particulates were multiplied by previously determined concentrations shown in table 4 (1) to yield the radioactivity emissions.

Table 1

Annual summary of emissions (a)

Ore drying facility

Source	Total Operating Time (hr)	Total Particulates g	Radium (b) ^{226}Ra μCi	Uranium (b) ^{234}U μCi	^{235}U μCi	^{238}U μCi	Thorium (b) ^{227}Th μCi	^{228}Th μCi	^{230}Th μCi	^{232}Th μCi
Phos Rock Dryer #1 (c)	4114	5.85×10^7	2450	2400	110	2400	120	40	2470	30
Phos Rock Dryers #3 & 4	4338	6.52×10^7	2740	2670	120	2670	130	40	2760	30
Phos Rock Transfer (combined totals for six stacks)	4338	3.76×10^7	1570	1540	70	1540	80	20	1590	20

(a) From 1975 Air Pollutant Emissions Report.
(b) Radioactivity results calculated from Facility Report and previous radioactivity measurements of phosphate rock.
(c) Combined totals for twin stacks.

Table 2

Annual summary of emissions (a)

Wet Process - Plant A

Source	Total Operating Time (hr)	Total Particulates g	Radium (b) ^{226}Ra μCi	Uranium (b) ^{234}U μCi	^{235}U μCi	^{238}U μCi	Thorium (b) ^{227}Th μCi	^{228}Th μCi	^{230}Th μCi	^{232}Th μCi
TSP Dryer	4560	4.9×10^5	12.5	34.5	1.7	34.5	0.71	0.54	28.7	0.77
Dry Product (TSP) Shipping	500	12.4×10^5	26.1	72.1	3.5	72.1	1.5	1.1	59.7	1.62
Phos Rock Grinding	3950	15.3×10^5	64.1	62.5	4.0	62.5	3.1	0.9	64.5	0.67
Phos Acid Process (Phos Rock)	6460	17.6×10^5	74	72.2	4.5	72.2	3.5	1.1	74.5	0.77
Phos Acid Process (Phos Rock)	4000	2.0×10^5	8.4	8.2	0.5	8.2	0.4	0.12	8.4	0.09

(a) From 1976 Air Pollutant Emissions Report.
(b) Radioactivity results calculated from Facility Report and previous radioactivity measurements of phosphate rock and GTSP.

Table 3

Annual summary of emissions (a)

Wet Process - Plant B

Source	Total Operating Time (hr)	Total Particulates g	Radium (b) ^{226}Ra μCi	Uranium (b) ^{234}U μCi	Uranium (b) ^{235}U μCi	Uranium (b) ^{238}U μCi	Thorium (b) ^{227}Th μCi	Thorium (b) ^{228}Th μCi	Thorium (b) ^{230}Th μCi	Thorium (b) ^{232}Th μCi
DAP Reactor/ Granulator	7516	7.19×10^7	403	4530	216	4530	115	58	4670	29
DAP Dryer	7516	7.41×10^7	415	4670	222	4670	119	59	4820	96
TSP Reactor/Blunger	7410	2.13×10^7	447	1240	60	1240	26	19	1020	28
TSP Dryer	7410	11.4×10^7	2390	6610	319	6610	140	102	5470	148
Product Storage (c)	8400	9.88×10^7	2070	5730	277	5730	119	89	4740	128
Product Shipping (c)	8400	11.7×10^7	2460	6790	328	6790	140	105	5620	150
Ore Unloading (Phos Rock)	8400	3.53×10^7	1480	1450	67	1450	71	22	1490	15
Unground Rock Storage Storage (5 sources - combined)	8400	1.50×10^7	630	615	29	615	30	9	630	7
Phos Rock Feed	8400	2.15×10^7	903	882	41	882	43	13	910	9
Phos Rock Grinding & Storage	8400	2.15×10^7	903	882	41	882	43	13	910	9
Ball Mill (Phos Rock)	8400	0.86×10^7	360	353	16	353	17	5	360	4
Ground Rock Silo	8400	2.04×10^7	860	840	39	840	41	12	863	9
Feed Bin (Phos Rock)	8400	1.52×10^{10}	640	620	29	620	30	9	640	7
Phos Rock Storage	8400	1.51×10^{10}	630	620	29	620	30	9	640	7

(a) From 1976 Air Pollutant Emissions Report.
(b) Radioactivity results calculated from Facility Report and previous radioactivity measurements of phosphate rock DAP and TSP.
(c) TSP radioactivity values used for calculations.

Table 4

Radium, uranium and thorium concentrations

in Florida phosphate industry products (a)

Material	Radium-226 (pCi/g)	Uranium (pCi/g)			Thorium (pCi/g)			
		234	235	238	227	228	230	232
Diammonium Phosphate (DAP)	5.6	63	3.0	63	1.6	0.8	65	0.4
Triple Super-Phosphate (TSP)	21	58	2.8	58	1.2	0.9	48	1.3
Marketable Phos Rock	42	41	1.9	41	2.0	0.61	42.3	0.44

(a) From reference (1).

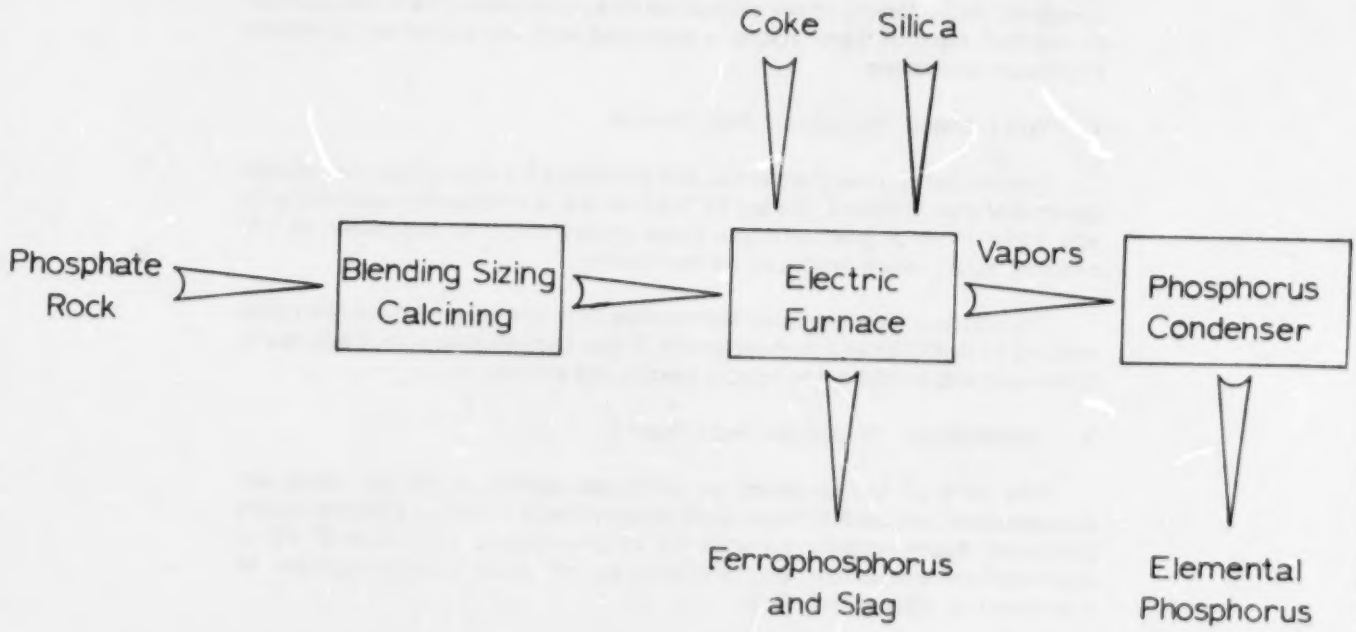

Figure 4. Thermal process flow diagram.

For example, the stack for dryers #3 and 4 at the ore drying facility released a total of 6.52×10^7 g of rock dust during 1976 and previous studies have shown that phosphate rock has a concentration of 42 pCi/g ^{226}Ra. Therefore, this stack released approximately (6.52×10^7 g x 42 pCi/g) 27.4×10^8 pCi of ^{226}Ra to the atmosphere during 1976. Similar calculations were made for each source, isotope and product.

1. Ore Drying Facility

The annual summary of emissions for the ore drying facility studied is shown in table 1. Dryer #1 vents through twin stacks 24 m tall, both 1.5 m in diameter. During 1976 this dryer processed 7.4×10^8 kg of wet phosphate rock. This dryer is equipped with a wet scrubber to reduce particulate emissions.

Dryers #3 and 4 vent through a common stack 24 m tall and 2.4 m in diameter. During 1976 these dryers processed a total of 1.98×10^9 kg of wet phosphate rock. These dryers are also equipped with wet scrubbers to reduce particulate emissions.

The six phosphate rock transfer stacks vary in height from 10.4 m to 47 m and in diameter from 0.7 m to 2.1 m. These stacks are used to vent the various points within the facility where phosphate rock is transferred from one location to another. Each of these stacks is equipped with wet scrubbers to reduce particulate emissions.

2. "Wet Process" Phosphoric Acid Plant A

The annual summary of particulate emissions for one of the wet process plants is shown in table 2. During 1976 a total of 4.08×10^8 kg of phosphoric acid and 2.27×10^8 kg of granular triple super phosphate, both expressed as 100 percent P_2O_5 were produced by this facility.

The emission sources listed in this table vent to the atmosphere at heights ranging from 42.7 to 24.4 m above grade. These stacks are equipped with fabric filters and wet scrubbers to reduce particulate emissions.

3. "Wet Process" Phosphoric Acid Plant B

The 1976 annual summary of particulate emissions for the other wet process plant included in this study is shown in table 3. During 1976 this facility produced approximately 4.31×10^8 kg of phosphoric acid, 2.0×10^8 kg of diammonium phosphate and 9.07×10^7 kg of triple superphosphate all expressed as 100 percent P_2O_5

The emission sources at this facility vent to the atmosphere at heights ranging from 12.2 m to 56.4 m. The stack diameters vary in diameter from 0.15 m to 2.4 m. Fabric filters and wet scrubbers are used to reduce particulate emissions from these sources.

B. EPA Field Study

1. Stack Sampling Data

This EPA field survey of the phosphate industry in central Florida consisted of two portions: *(1) measurement of actual particulate emissions from selected stacks at each facility; and (2) ambient air samples collected in the immediate vicinity of each plant and other locations in the general area of the phosphate milling operations.*

Stack measurements were performed with a particulate sampling train similar to the one shown in figure 5 and in accordance with EPA guidelines set forth in the Code of Federal Regulations, Title 40, Part 60 (5). The sampling train uses a 65 mm glass fiber filter to trap the particulates removed from the stack via the sampling probe. Each filter was preweighed prior to sampling and following the sampling period the net filter weight was used to determine the total particulate catch. After the particulate weight had been determined, the filters were boiled with acid to remove particulate matter. Radiochemical analyses were then used to determine the amount of each specific radioisotope. A total of seven stacks were sampled by this method, two at the ore drying facility and five at the two wet process phosphoric acid plants. Resource constraints did not permit the sampling of all emission sources within each facility. The emission sources sampled were selected based on previous sampling data by each facility and operations known to produce radioactive effluents. The EPA survey data were used to supplement the facility reported data.

The results of the EPA stack sampling survey are shown in table 5. The annual summary of emissions for the sources sampled during the EPA survey are shown in table 6. This summary is based on the operating times for 1976 and the survey results given in table 5.

The EPA results are compared to the facility data in table 7. Only six of the sampled stacks are shown in this table because the TSP dryer at plant A actually has two vents to the atmosphere, one at the 30 m level and the other at the 42 m level. The facility normally only samples the 30 m vent for particulates and 42 m vent for fluoride. Facility reported data in table 2 reflects particulate emissions at the 30 m level only. During the EPA survey only particulates being emitted from the 42 m level were sampled. Therefore, to obtain the total particulate emissions for this dryer, it is necessary to sum the results for the two levels.

Generally speaking, good agreement is noted between EPA data and some of the facility reported data. Dryers #3 and 4 reported releases are in excellent agreement with the EPA results. The facility operator at the ore dryer facility stated that substantial modifications had been performed on the wet scrubber for dryer # 1 which should have reduced the total emissions from these twin stacks. In all cases the wet process plant B reported emissions were greater than those measured by EPA. In some instances the plant data is higher by a factor of 10. This overestimation of releases by the facility operator will in turn result in an overestimation of radiation doses.

Stack	Hours Per Year	Stack Flow m^3/min	Vol. Sampled m^3	Sample Weight mg	Ra-226 pCi/filter	U-234 pCi/filter	U-235 pCi/filter	U-238 pCi/filter	Th-227 pCi/filter	Th-228 pCi/filter	Th-230 pCi/filter	Th-232 pCi/filter
Rock Dryer #1	4114	1280	1.76	59.3	2.6 ± 3%	1.95 ± 16%	.084 ± 50%	1.84 ± 16%	.136 ± 52%	.403 ± 20%	2.69 ± 8%	.074 ± 49%
Rock Dryers #3 & 4	4338	5390	2.68	95.3	5.6 ± 2%	5.01 ± 13%	.45 ± 23%	5.21 ± 13%	.39 ± 33%	.436 ± 22%	4.75 ± 6%	.152 ± 37%
TSP Dryer Plant "A"	4560	3100	1.65	38.7	.06 ± 12%	.214 ± 28%	ND	.176 ± 31%	ND	.062 ± 48%	.172 ± 32%	ND
TSP Dryer Plant "B"	7411	2378	1.93	20.6	.40 ± 7%	.545 ± 21%	.034 ± 73%	.50 ± 22%	ND	.047 ± 60%	.453 ± 19%	.134 ± 36%
TSP R/BL Plant "B"	7411	136	1.64	7.0	.12 ± 11%	.187 ± 31%	ND	.164 ± 33%	ND	.078 ± 49%	.16 ± 34%	ND
DAP Dryer Plant "B"	7516	2200	1.32	20.1	.34 ± 8%	4.74 ± 7%	.248 ± 28%	4.37 ± 7%	ND	.109 ± 41%	3.94 ± 7%	.068 ± 52%
DAP R/G Plant "B"	7516	788	1.39	27.0	.04 ± 26%	2.42 ± 10%	.141 ± 38%	2.18 ± 10%	ND	.113 ± 40%	1.15 ± 13%	.036 ± 73%

ND - Non detectable.

Table 5

EPA Stack Sampling Data

Table 6

Annual summary of emissions for sources sampled (a)

by EPA

Source	Total Particulates g	^{226}Ra μCi	Uranium ^{234}U μCi	Uranium ^{235}U μCi	Uranium ^{238}U μCi	Thorium ^{227}Th μCi	Thorium ^{228}Th μCi	Thorium ^{230}Th μCi	Thorium ^{232}Th μCi
Rock Dryer #1 (b)	2.2×10^7	930	700	30	660	50	140	97	30
Rock Dryers #3 & 4	5.0×10^7	2900	2600	240	2700	200	230	2490	80
TSP Dryer Plant "A"	2.0×10^7	30	110	ND	90	ND	40	90	ND
TSP Dryer Plant "B"	1.2×10^7	220	300	20	270	ND	30	250	70
TSP R/BL Plant "B"	0.2×10^7	4	7	ND	7	ND	3	6	ND
DAP Dryer Plant "B"	1.5×10^7	260	2560	190	3280	ND	80	2960	50
DAP R/G Plant "B"	0.7×10^7	10	620	40	560	ND	30	290	9

(a) Based on EPA sampling data and operating times for 1976.

(b) Combined totals for twin stacks.

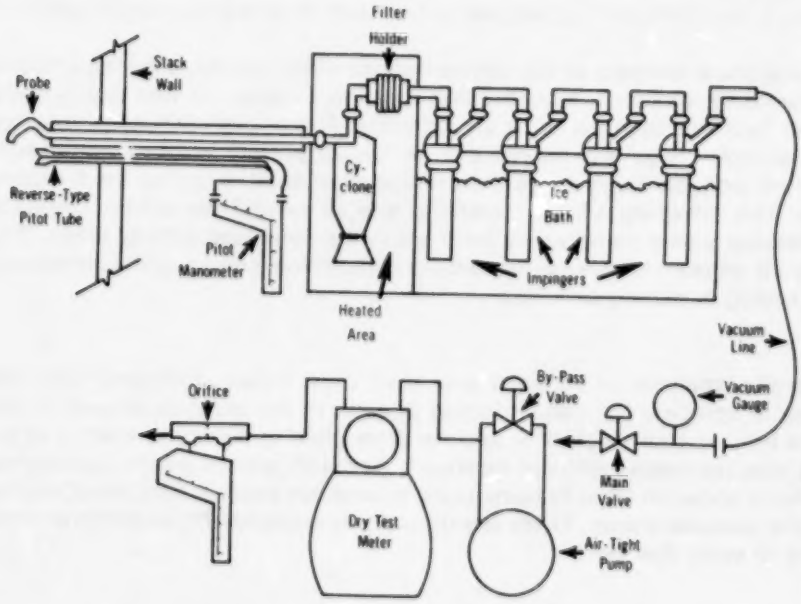

Figure 5. EPA stack sampling train.

In addition to the stack sampling, dust samples were collected from the TSP bag house and from the ball mill dust collector at the wet process plant A. These samples were returned to the laboratory for ^{210}Po analysis. Based on these analyses, total annual ^{210}Po emissions for plant A were calculated and are shown in table 8.

2. Off-Site Air Sampling Data

High-volume air samplers were used to collect particulate samples around the two wet process plants and the ore drying plant. Each sampler utilized a 10 cm respirator type filter designed to trap dusts (MSA-BM-2133). Typical flow rates ranged from 1416 l/m (50 cfm) to 1700 l/m (60 cfm) at the start to 1133 l/m (40 cfm) to 1416 l/m (50 cfm) after 2 to 3 days operation. Each sampler was placed in a wooden housing mounted approximately 1 m above ground to protect it from weather effects and ground level dust.

Locations for air samplers were selected primarily by availability of electrical power at distances of approximately 300 to 1000 m from each plant. Whenever possible samples were collected in the plume downwind from the plants where stack sampling was being done. Distance and direction from each plant for each sampling location are provided in tables 9 and 10. Tables 9 and 10 also indicate the volume of air drawn through the filter and the activities of ^{226}Ra, ^{234}U, ^{235}U, ^{238}U, ^{227}Th, ^{228}Th, ^{230}Th, and ^{232}Th per cubic meter of air for each location.

Ambient particulate levels for the general area were determined by operating air samplers at seven additional sites (see figures 6 and 7.) These were located coincident with Florida State Department of Environmental Regulation air monitoring sites in Polk County. These samplers were operated for two periods of 48 to 72 hours. Sample volume and activities of radium, uranium, and thorium isotopes per cubic meter of air are provided in table 11.

Statistical analysis of the concentrations of each isotope for the ambient air sampling locations indicates that location I (table 11) had significantly higher activities than the other six locations. A one-way analysis of variance and multiple range test proved that, at the 95 percent level of confidence, ambient location I was above the average level determined for the six other sites. The proximity of this sampling site to several phosphate chemical processing plants could easily have led to the increased activity levels. The other six ambient sites were much more distant from any on-going phosphate processing or mining activities.

One-way analysis of variance was then used (when sufficient data was available) to compare the concentration of each of the isotopes at each of the locations around plant A, plant B, and the dryer plant to the concentration at the ambient sites (excluding ambient location I). At the 95 percent level of confidence the locations shown in table 12 were found to be above ambient concentrations for each of the isotopes shown. There was not sufficient data for ^{235}U and ^{227}Th at these locations to apply this test.

Table 7

Comparison of EPA and facility sampling data (a)

	Total Particulates		Radium-226		Uranium-234		Thorium-230	
	Facility Data g	EPA Data g	Facility Data μ Ci	EPA Data μ Ci	Facility Data μ Ci	EPA Data μ Ci	Facility Data μ Ci	EPA Data μ Ci
Rock Dryer #1	5.85×10^7	2.2×10^7	2450	930	2400	700	2470	970
Rock Dryers #3 & 4	6.52×10^7	5.0×10^7	2740	2900	2670	2600	2760	2500
TSP Dryer "B"	11.4×10^7	1.2×10^7	2390	220	6610	300	5470	250
TSP R/BL "B"	2.13×10^7	0.2×10^7	447	4	1240	7	1020	6
DAP Dryer "B"	7.41×10^7	1.5×10^7	400	260	4600	2560	4800	3000
DAP R/G "B"	7.19×10^7	0.7×10^7	400	10	4500	620	4600	300

(a) Annual summary based on 1976 operating times.

Table 8

^{210}Po

Wet process plant A (a)

Source	Total Particulates g	^{210}Po μ Ci/yr
TSP Dryer	200×10^5 (b)	630
Dry Product (TSP) Shipping	12.4×10^5	39
Rock Grinding	15.3×10^5	62
Phos Acid Process (Rock)	17.6×10^5	71
Phos Acid Process (Rock)	2.0×10^5	8

(a) Based on a concentration of 31.6 pCi/g ^{210}Po in TSP and 40.3 pCi/g ^{210}Po in phos rock.

(b) Sum of two stack vents (100' and 140').

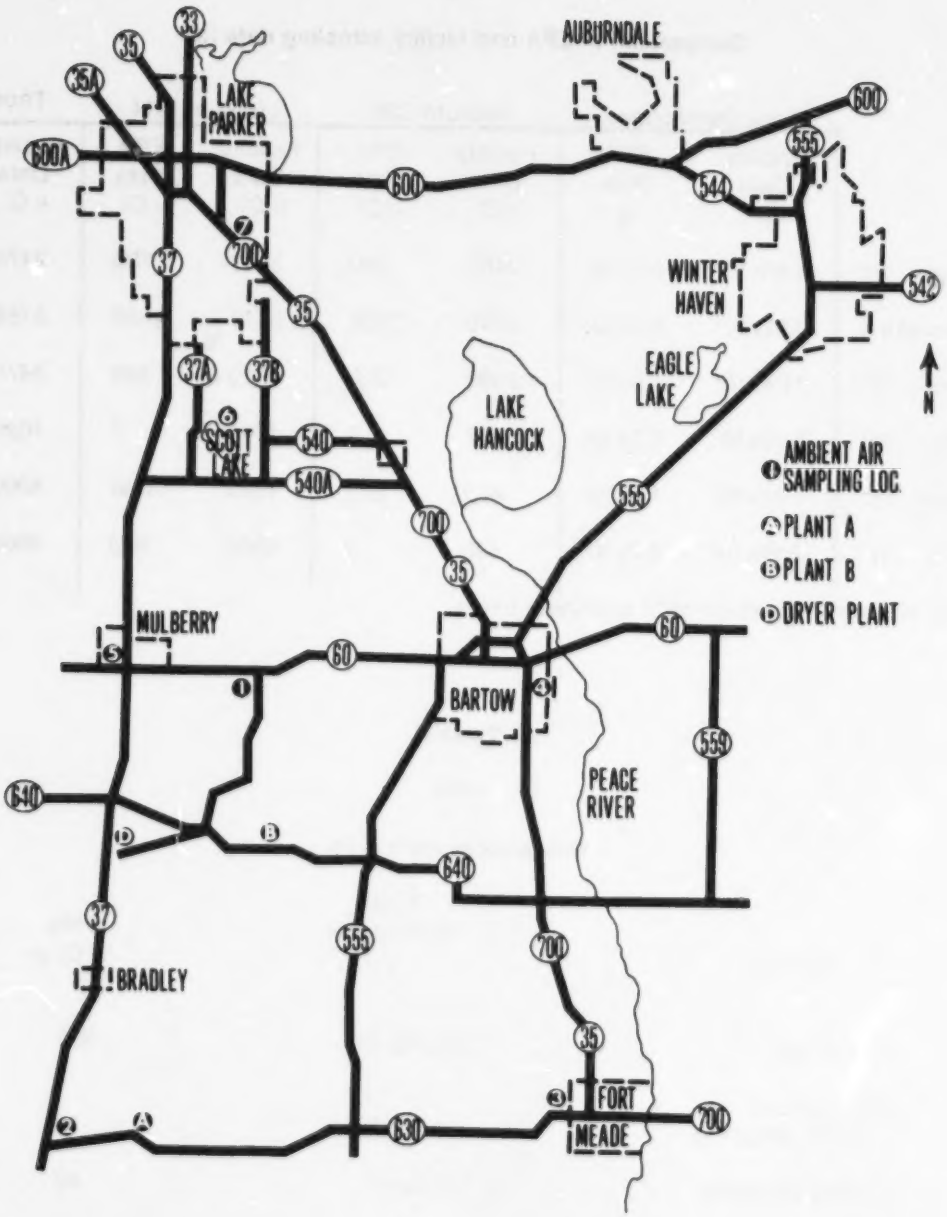

Figure 6. Off-site air sampling sites.

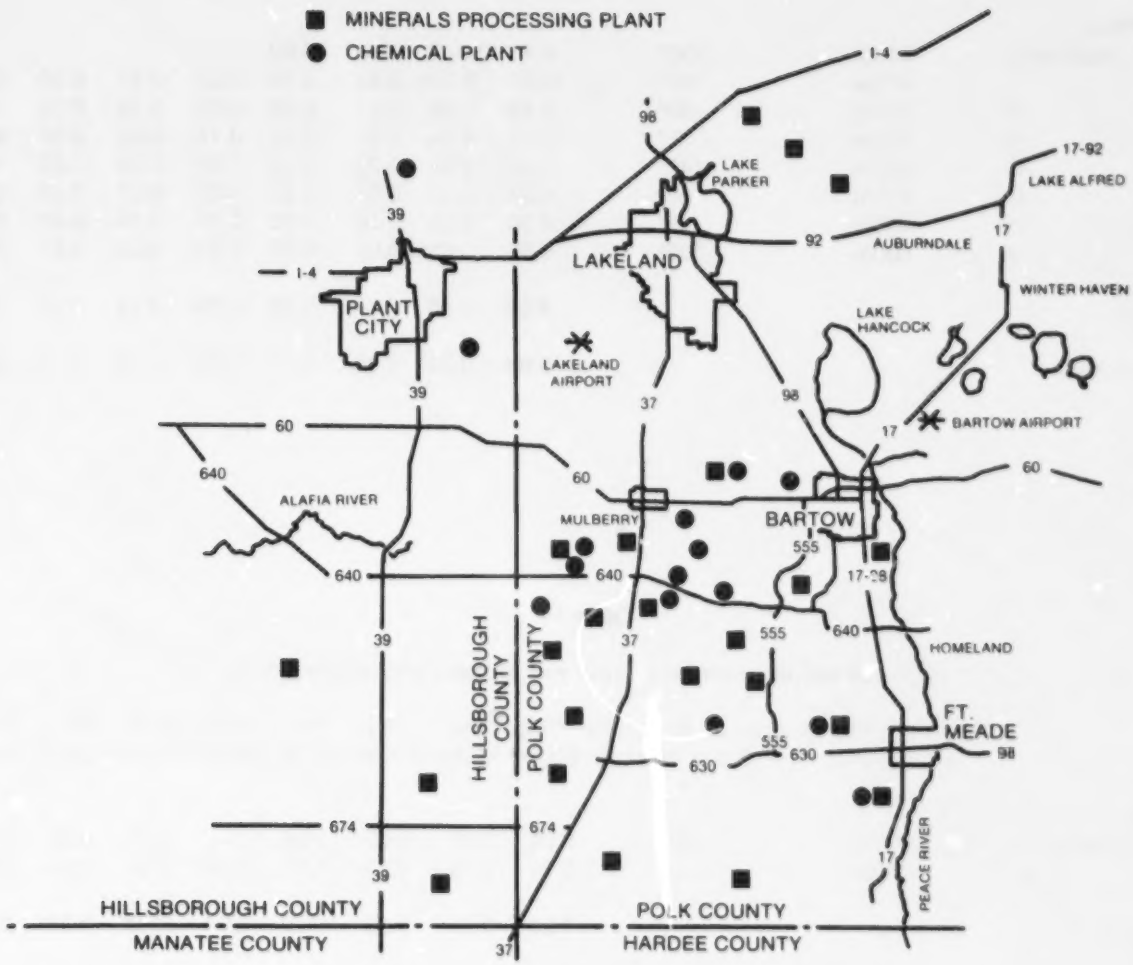

Figure 7. Phosphate mining and processing area.

Table 9

High volume air samples around phosphate ore dryer plant

	Approx. Distance From Plant	Approx. Direction From Plant	^{226}Ra fCi/m³	^{234}U fCi/m³	^{235}U fCi/m³	^{238}U fCi/m³	^{227}U fCi/m³	^{228}Th fCi/m³	^{230}Th fCi/m³	^{232}Th fCi/m³
Dryer Plant										
Location I	260m	230°	6.41	19.20	0.90	18.90	-	-	-	-
II	400m	140°	10.50	8.92	0.48	8.58	0.33	0.41	8.36	0.46
III	375m	90°	0.49	0.56	0.01	0.52	0.05	0.13	0.76	0.07
IV	400m	35°	5.13	4.39	0.21	4.35	0.17	0.22	4.57	0.07
II	400m	140°	8.04	4.68	0.20	4.60	0.30	0.28	7.20	0.19
III	375m	90°	1.24	1.16	0.06	1.20	0.05	0.07	1.20	0.04
IV	400m	35°	4.20	3.51	0.18	3.51	0.11	0.10	3.41	0.10
V	400m	335°	0.67	0.65	0.02	0.66	0.03	0.06	0.61	0.04
Average			4.59	5.38	0.26	5.29	0.15	0.18	3.73	0.14
Std. Deviation			3.66	6.23	0.30	6.11	0.12	0.13	3.14	0.15

Table 10

High volume air samples around wet process phosphate plants

	Approx. Distance From Plant	Approx. Direction From Plant	^{226}Ra fCi/m³	^{234}U fCi/m³	^{235}U fCi/m³	^{238}U fCi/m³	^{227}Th fCi/m³	^{228}Th fCi/m³	^{230}Th fCi/m³	^{232}Th fCi/m³
Plant A										
Location I	1000m	65°	0.26	0.25	0.03	0.24	-	0.02	0.31	0.01
II	750m	145°	2.39	0.68	0.03	0.66	0.04	0.04	0.67	0.03
Average			1.33	0.47	0.03	0.45	0.04	0.03	0.49	0.02
Std. Deviation			1.51	0.30	-	0.30	-	0.01	0.25	0.01
Plant B										
Location I	1000m	100°	0.41	0.53	0.03	0.53	0.03	0.09	0.60	0.03
II	300m	210°	1.91	2.22	0.09	2.38	-	0.44	2.74	0.16
III	1370m	275°	0.09	0.11	-	0.12	-	0.02	0.16	0.02
IV	2100m	275°	0.21	0.32	0.01	0.28	0.01	0.07	0.26	0.04
V	1000m	100°	1.31	1.31	0.06	1.34	0.07	0.12	1.42	0.04
Average			0.786	0.898	0.05	0.93	0.04	0.148	1.04	0.06
Std. Deviation			0.789	0.867	0.04	0.94	0.03	0.167	1.07	0.06

Table 11

Ambient air sampling stations

Location	^{226}Ra fCi/m³	^{234}U fCi/m³	^{235}U fCi/m³	^{238}U fCi/m³	^{227}Th fCi/m³	^{228}Th fCi/m³	^{230}Th fCi/m³	^{232}Th fCi/m³
Ambient Station I	5.64	6.78	0.31	6.82	0.20	0.18	5.85	0.25
I	2.60	2.74	0.12	2.72	0.11	0.08	2.64	0.07
II	0.43	0.48	-	0.49	-	0.08	0.64	0.07
II	1.41	0.82	0.04	0.75	0.05	0.04	0.78	0.04
III	1.22	1.01	0.07	1.05	0.09	0.26	0.96	0.14
III	0.77	0.61	0.03	0.68	0.06	0.18	0.69	0.13
IV	0.13	0.11	-	0.10	-	0.07	0.12	0.02
IV	0.12	0.06	-	0.07	-	0.06	0.08	0.01
V	0.28	0.23	0.01	0.23	0.02	0.77	0.31	0.02
V	0.73	0.83	0.03	0.77	0.05	0.09	0.81	0.03
VI	0.20	0.17	0.01	0.17	0.03	0.08	0.17	0.03
VI	0.12	0.07	-	0.07	-	0.10	0.08	0.02
VII	0.17	0.14	0.01	0.14	-	0.03	0.18	0.02
VII	0.21	0.16	-	0.16	-	0.04	0.18	0.03
Average	1.00	1.02	0.07	1.02	0.08	0.15	0.96	0.06
Std. Deviation	1.51	1.80	0.10	1.81	0.06	0.19	1.56	0.07
Locations II-VII Average	0.48	0.39	0.03	0.39	0.05	0.15	0.42	0.05
Std. Deviation	0.45	0.34	0.02	0.34	0.02	0.20	0.33	0.04

Table 12

Isotopes above average ambient concentration

Location	Isotopes
Plant A	
Location II	^{226}Ra
Plant B	
Location II	^{226}Ra, ^{234}U, ^{238}U, ^{228}Th, ^{230}Th, ^{232}Th
Dryer Plant	
Location I	^{226}Ra, ^{234}U, ^{238}U
Location II	^{226}Ra, ^{234}U, ^{238}U, ^{230}Th, ^{232}Th
Location IV	^{226}Ra, ^{234}U, ^{238}U, ^{230}Th

Particle size analysis of the air in the plume from stacks at wet process plant A and the ore dryer plant were done with Andersen 2000 Air Samplers (Model 65-000). These samplers utilized a series of offset filters in a specially designed system plate that sizes aerodynamically suspended particulate matter into four fractionations (1.1, 2.0, 3.3, and 7.0 μm) with the submicron material being trapped on a backup filter. High-volume air samplers used with the Andersen 2000 samplers were calibrated with a water-filled manometer to provide 566 liters per minute (20 cfm) flow through the filter system. These samplers were placed directly in the plume from the plant with the aid of portable electric generators. The results of the analysis of these samples are shown in table 13.

A log normal distribution was assumed for the activities deposited on the Andersen filters. For each Andersen sampler run ^{234}U, ^{226}Ra, and ^{230}Th activities were plotted on log probability paper (figures 8, 9, and 10) to determine the AMAD (activity median aerodynamic diameter) of the particulate emissions. For the dryer plant a value of 8.2 μm was found for the AMAD while the AMAD value for the wet process plant B was 2.8 μm for the first run and 2.9 μm for the second run. Based on the preceding results, a median particle size of 8 μm was assumed for dryer plant emissions and an AMAD of 3 μm was assumed as the median diameter of particulate emissions from both wet process plants.

IV. Dose Assessment

The computer code AIREM (6) was used to make dose estimates resulting from plant emissions given in tables 1, 2, 3, and 6. AIREM uses a sector averaged diffusion equation to determine an average concentration or dose in a given sector at a specific downwind distance. The lung dose conversion factors are derived from information contained in the ICRP Task Group Lung Model Report (7) and ICRP Report Number 19 (8). A list of assumptions, dose conversion factors, and total source terms for each plant are given in tables 14, 15, 16, 17, and 18. The total source terms were based on the reported operating times for 1976 and, where available, EPA stack sampling results were used. In those cases where EPA data were not available, previously reported data given in tables 1, 2, and 3 were used.

Individual and population dose estimates to the lung were made for each plant. Individual dose estimates were based on the nearest residence to each plant except in the case of wet process plant A where no residence was nearby. Individual dose estimates were also made for the maximum lung dose at the 400 m distance. This distance represents the nearest residence assumed to be realistically possible at a typical plant where the distance is measured from the point of release and not the site boundary. This distance was chosen as a reference point for comparison purposes only. Population doses were based on population distributions generated by computer code from U.S. Census Bureau information for this area. Individual lung dose estimates (mrem/yr) and population doses (person-rem/yr) are given in tables 19 and 20. Ground level release with deposition and depletion plus a building wake correction (9) were assumed for the individual dose calculations. Elevated release points were assumed for some calculations; however, they did not significantly change the results. The results of elevated release point calculations are not shown in this report.

The dose estimates from wet process plant A reflect the addition of ^{210}Po to the source term as shown in table 8. These ^{210}Po values were available for this plant only and are estimated to contribute approximately 12 percent of the annual dose from wet process plant A releases.

Table 13

Andersen samplers operated at Polk County, FL

(fCi/m³)

Particle Size	7 μm	3.3-7.0 μm	2.0-3.3 μm	1.1-2.0 μm	1.1 μm
Location/Isotope	Filter A	Filter B	Filter C	Filter D	Filter E
Ore Dryer					
^{234}U	21.9	9.33	3.74	2.56	4.98
^{235}U	1.46	-	-	-	0.618
^{238}U	21.1	8.95	4.03	2.46	5.41
^{227}Th	-	-	-	-	-
^{228}Th	1.13	1.34	0.96	1.09	0.982
^{230}Th	22.7	10.8	4.96	2.56	5.70
^{232}Th	0.541	1.00	-	-	-
^{226}Ra	26.5	9.27	4.64	4.42	5.96
Sample Gross Wt.	0.033g	0.014g	0.006g	0.003g	0.006g
Wet Process Plant B					
Run 1					
^{234}U	2.54	1.18	1.04	1.08	1.70
^{235}U	-	-	-	-	-
^{238}U	1.85	0.850	0.794	1.26	1.95
^{227}Th	-	-	-	-	-
^{228}Th	1.14	0.784	1.04	0.993	0.960
^{230}Th	3.12	3.18	3.94	0.993	0.960
^{232}Th	0.706	-	0.828	-	0.861
^{226}Ra	1.77	2.43	2.87	2.65	2.87
Sample Gross Wt.	0.018g	0.0004g	0.0004g	-	-
Run 2					
^{234}U	0.985	1.44	1.08	0.364	1.04
^{235}U	-	0.175	0.275	-	-
^{238}U	1.49	1.44	1.32	0.364	0.926
^{227}Th	-	-	-	-	-
^{228}Th	0.286	0.372	0.335	0.282	0.283
^{230}Th	1.24	0.810	1.06	0.870	1.11
^{232}Th	0.175	-	0.335	-	0.632
^{226}Ra	1.12	0.743	0.892	0.818	0.967
Sample Gross Wt.	0.002g	0.015g	-	0.0008g	0.005g

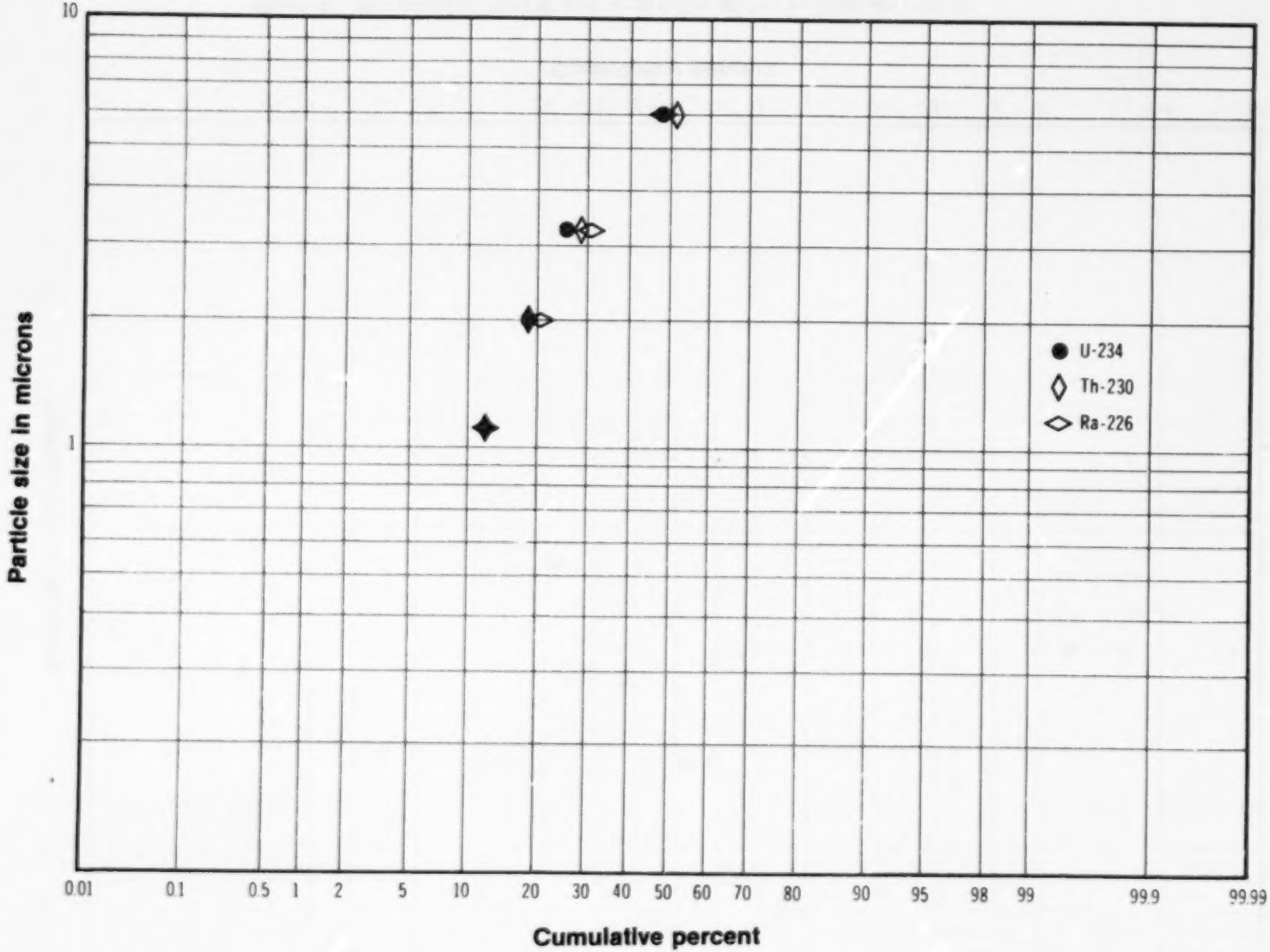

Figure 8. Ore drying plant. Log probability plot of particle size.

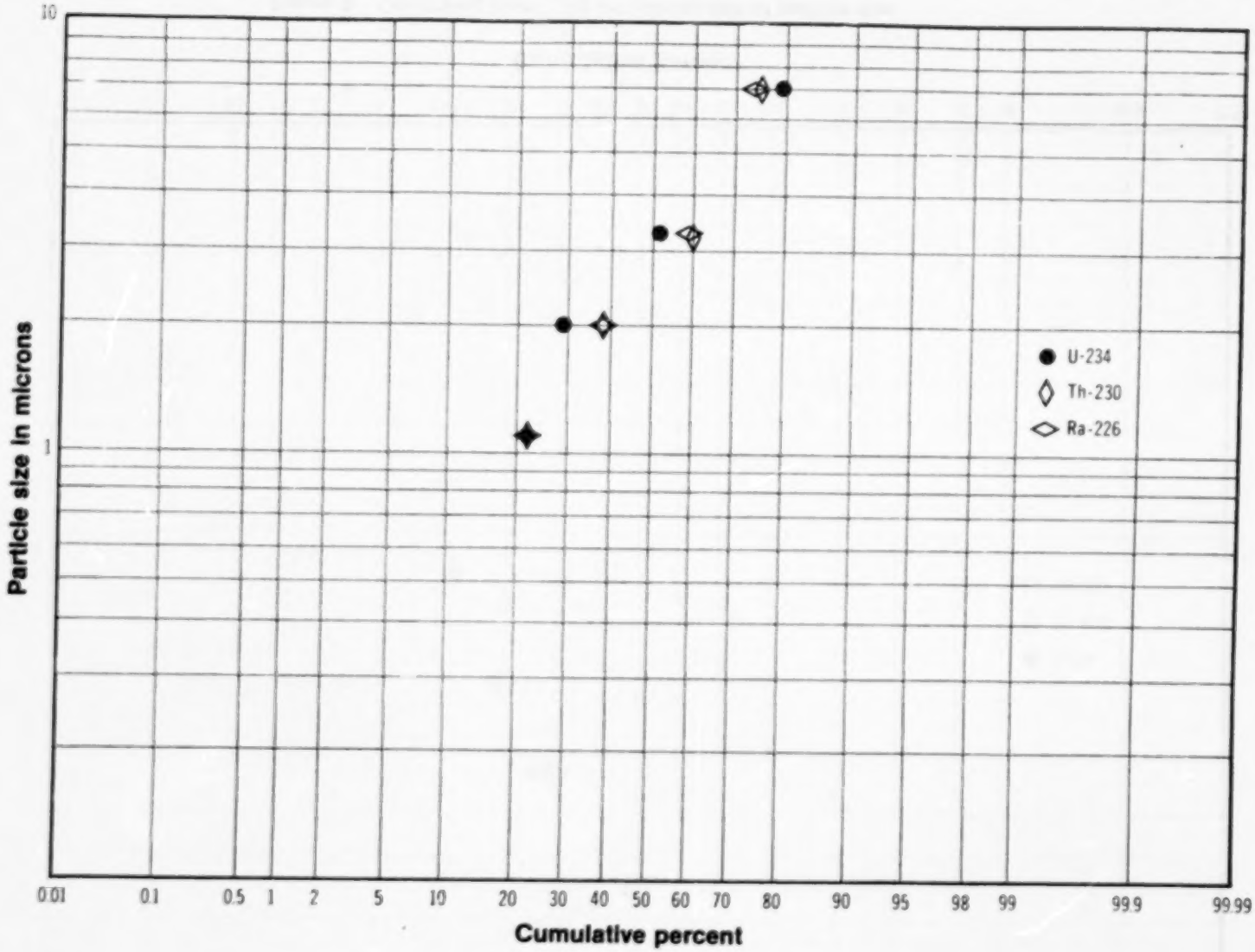

Figure 9. Wet process Plant B. Log probability plot of particle size run #1.

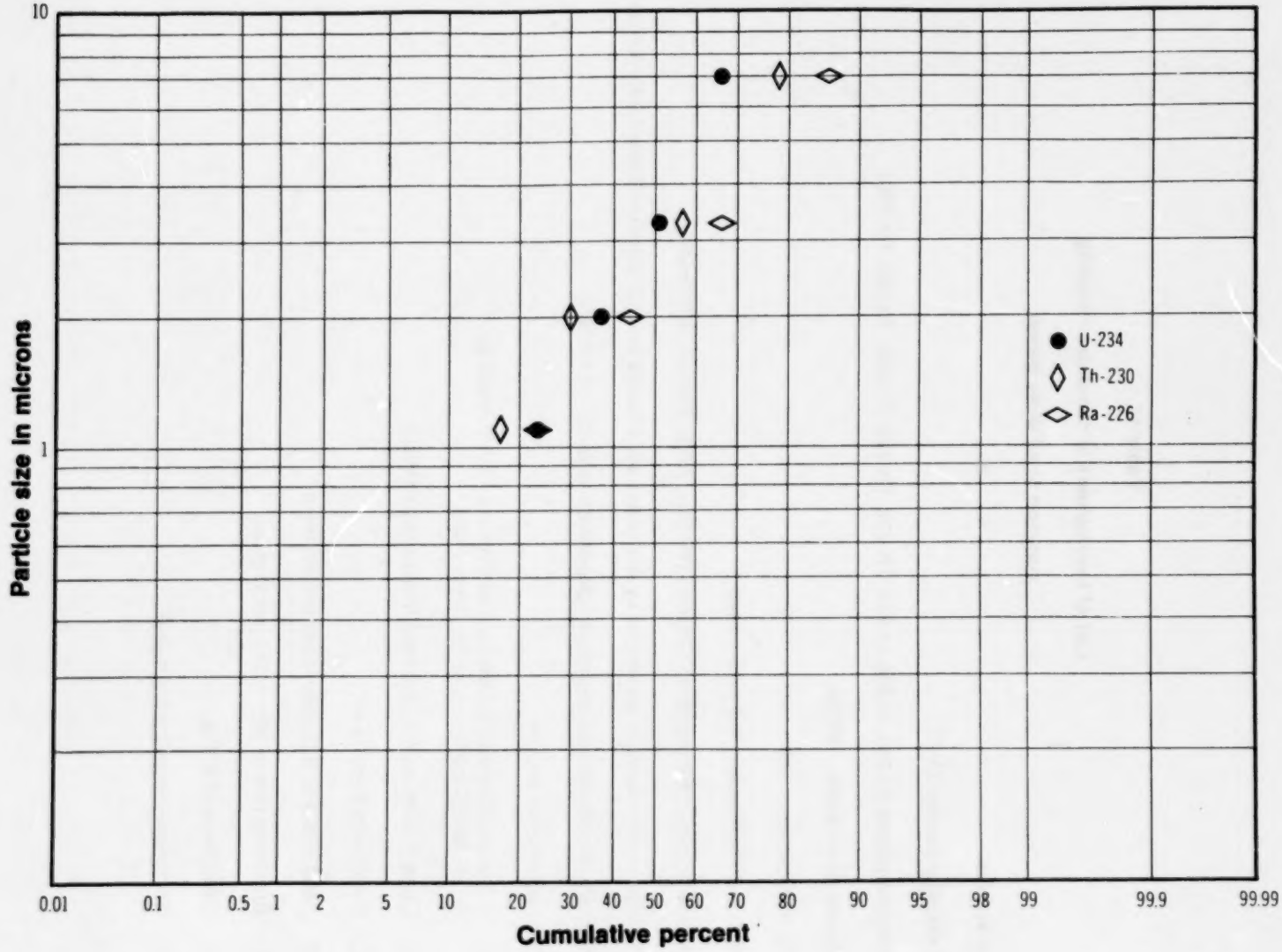

Figure 10. Wet process Plant B. Log probability plot of particle size run #2.

Table 14

List of assumptions in computer modeling

common to all three plants

1. 16 sectors
2. 5 stability classes (A-E)
3. 8 radionuclides (U-238, U-235, U-234, Th-227, Th-228, Th-230, Th-232, Ra-226)
4. Mixing layer depth: 1000m
5. Rainfall fraction: .05
6. Washout coefficient: 2.0×10^{-4} l/sec
7. Meteorological data based on Orlando (McCoy, AFB), Florida, information
8. Population distributions generated by computer code based on U.S. Census Bureau information
9. With dry deposition and depletion - deposition velocity: 1 cm/sec
10. Dose conversion factors
    - A. 3 m AMAD particle size for Wet Process Plants A and B
      8 m AMAD particle size for Dryer Plant
    - B. Class Y lung model (assumes insoluble particles)
    - C. Pulmonary lung dose
    - D. Dose refers to a 50 year dose commitment
    - E. Breathing rate of 23 m^3/day (adult male)
    - F. Lung mass of 570 g
    - G. Continuous expsure for a year

Table 15

Dose conversion factors

Isotope	(mrem/sec)/(Ci/m³)
^{226}Ra	1.20×10^{8}
^{238}U	1.02×10^{8}
^{235}U	1.10×10^{8}
^{234}U	1.20×10^{8}
^{227}Th	2.80×10^{7}
^{228}Th	4.60×10^{8}
^{230}Th	1.10×10^{8}
^{232}Th	1.60×10^{8}
^{210}Po	2.80×10^{7}

Table 16

Dryer plant source term*

Isotope	Ci/year
^{238}U	4.90×10^{-3}
^{235}U	3.41×10^{-4}
^{234}U	4.84×10^{-3}
^{227}Th	3.25×10^{-4}
^{228}Th	3.93×10^{-4}
^{230}Th	5.05×10^{-3}
^{232}Th	1.27×10^{-4}
^{226}Ra	5.40×10^{-3}

*Based on operating times given in table 1.

Table 17

Wet process plant A source term*

Isotope	Ci/year
^{238}U	3.40×10^{-4}
^{235}U	1.40×10^{-5}
^{234}U	3.60×10^{-4}
^{227}Th	9.20×10^{-6}
^{228}Th	4.38×10^{-5}
^{230}Th	3.26×10^{-4}
^{232}Th	3.90×10^{-6}
^{226}Ra	2.15×10^{-4}
^{210}Po	8.10×10^{-4}

*Based on operating times given in table 2.

Table 18

Wet process plant B source term*

Isotope	Ci/year
^{238}U	2.3×10^{-2}
^{235}U	1.1×10^{-3}
^{234}U	2.3×10^{-2}
^{227}Th	6.0×10^{-4}
^{228}Th	4.3×10^{-4}
^{230}Th	2.0×10^{-2}
^{232}Th	4.8×10^{-4}
^{226}Ra	1.1×10^{-2}

*Based on operating times given in table 3.

Table 19

Individual dose
(mrem/yr)

Source	Maximum		Nearest Residence	
	Location	Lung Dose	Location	Lung Dose
Dryer Plant	400m S	15	400m NNW	5.5
			840m S	7.4
Wet Plant "A"	400m S	1.4	800m W*	0.7
Wet Plant "B"	400m S	60	1860m W	5
			2700m E	1.5

* No residence at this location. The nearest residence would receive much less than 1 mrem/yr.

Table 20

Population doses within 80 km (a)

Source	Person-rem/yr (b)
Dryer Plant	1.2
Wet Plant A	0.1
Wet Plant B	6.6

(a) Ground level release assumed

(b) To the lung

Table 21

Dose predictions based on high volume samples (a)

Location	Lung Dose (mrem/yr)
Average Ambient w/o Location I	8.5 (b)
Ambient Station I	33.0 (c)
Wet Process Plant B Location II (300 m, 210°)	21.0 (c)

(a) Based on data in tables 10 and 11.

(b) 1 m AMAD particle size.

(c) 3 m AMAD particle size.

V. Summary and Conclusions

The results of this study show small, but measurable, increases in levels of radioactivity in air surrounding these selected phosphate milling operations. This is evidenced by results of the air sampler measurements shown in tables 9, 10, 11, and 12. However, it should be noted that levels statistically above background were measured in only 5 of the 12 locations sampled. Also these locations were all within 750 m from the respective plants. The dose estimates based on data from ambient locations also show the effects of the phosphate industry airborne emission. The projected lung dose at ambient location I is 33 mrem/yr compared with an average of 8.5 mrem/yr at the remaining ambient locations.

Dose projections based on stack release data also indicate the magnitude of individual lung doses in the area. The maximum individual dose at the nearest actual residence is estimated to be 7.4 mrem/yr (above background, i.e., in addition to background). This location was 840 m south of the ore drying facility.

Population doses within 80 km of the wet process plant B were calculated to be 6.6 person-rem per year. However, this dose is most likely overestimated due to the overestimation of releases by the facility operator.

The estimated doses based on stack sampling data and on high-volume sampling data at location II near wet process plant B were within a factor of 4 and most likely would be closer if more accurate source terms were available.

In conclusion, the results of this study show slight increases in the levels of radioactivity in air surrounding the plants studied. These slight increases in air concentration are estimated to produce an individual lung dose of a few mrem/yr to persons living in the immediate area of these plants. These estimations are based on stack measurements at the point of release and on air samples collected at the point of interest. Based on our data, it appears the ore drying operations are the most significant source of airborne radioactive particulates.

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